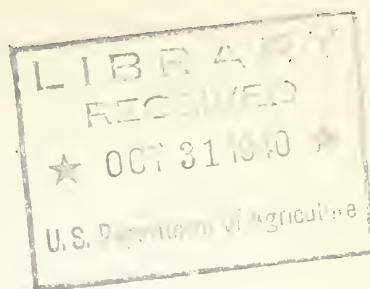


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Soybean Protein

RESUME AND BIBLIOGRAPHY

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U. S. Department of Agriculture
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SOYBEAN PROTEIN--RESUME AND BIBLIOGRAPHY

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INTRODUCTION

Until recently in the United States, the primary interest in the soybean was directed to its use as a feedstuff, especially for stock and poultry. It was soon found that the oil extracted from the soybean possessed properties of value for industrial application. Consequently the beans were processed by mechanical pressure or solvent extraction to remove the oil as completely as practical, and the meal was used as a protein concentrate in feeds. Within the past 5 years, however, the soybean acreage has steadily increased until now (1938) it approaches 7,000,000 acres (7) 2/ which yield about 50,000,000 bushels (89). Certain industrial uses have been developed for the meal and protein (78) extracted from the beans. In most cases these uses capitalize on the chemical characteristics of the protein and the relatively high percentage of protein found in soybean meal.

In view of this interest it seems appropriate to review the literature on soybean protein with special emphasis on its industrial applications. This is in no sense a complete review of all the work that has been done on soybean protein, but an effort has been made to cover enough of the field to give a fair picture of its development, to indicate present trends, and to suggest possible lines of research that appear most promising for the future. The use of soybeans in the field of nutrition has not been included except for a few references on the determination or detection of soybean flour in mixtures (2, 26, 28, 40, 45, 46, 60, 61, 76, 85, 121), and the agronomic aspect has been treated only in an introductory way. For other bibliographies on the general subject of "Soybeans," reference is made to the following:

Bibliography on Soybeans. Seattle Public Library,
Technology Division, 1932. (Out of print.)

The Composition and Nutritive Properties of Soybeans
and Soybean Oil Meal: A Literature Review. Prepared
by Soybean Nutritional Research Council, Chicago,
Illinois. October 1938.

1/ A cooperative organization participated in by the Bureaus of Agricultural Chemistry and Engineering and Plant Industry of the U. S. Department of Agriculture, and the Agricultural Experiment Stations of the North Central States of Illinois, Indiana, Iowa, Kansas, Michigan, Minnesota, Missouri, Nebraska, North Dakota, Ohio, South Dakota, and Wisconsin.

2/ Numbers in parentheses refer to Literature Cited, page 20.

Partial List of References on Soybeans and Soybean Products. U. S. Department of Agriculture, Bureau of Chemistry and Soils, Food Research Division. 1933. (Mimeographed.)

The Soybean. By C. V. Piper and W. J. Morse, pp. 288-310, McGraw-Hill, New York, 1923.

The Soybean Industry. Compiled by H. E. Hennefrund and E. M. Colvin, U. S. Department of Agriculture, Bureau of Agricultural Economics Bibliography No. 74. 1938. (Mimeographed.)

Soybean Industry in the United States. Compiled by M. I. Herb. U. S. Department of Agriculture, Bureau of Agricultural Economics. 1931. (Mimeographed.)

Soybeans, A Partial Bibliography. Compiled by E. I. Miller. Tennessee Valley Authority Technical Library. 1935. (Mimeographed.)

AGRONOMIC AND GENERAL

The soybean, which probably originated in Manchuria (87) and Japan, has attracted world-wide interest and is being widely cultivated. The 2,000 or more varieties adapt it to practically every climate suitable to vegetation, as indicated by its cultivation in Manchuria (51, 90), Austria (32), British Guiana and Jamaica (5, 65), Malay States (101), Dutch Indies (8), China (63), South Australia (111), Tasmania (37), Philippine Islands (91), South Africa (94, 95, 110), Belgian Congo (62), Rumania (108), Czechoslovakia (47, 102), Germany (38), Russia (53, 100), France (16, 67, 93), Italy (109), England (20, 86), Canada (10, 30, 31, 64, 98), and other countries. A great deal of work has been done in the United States within the last 5 years on the agronomic development of soybeans, and a study of the microscopic structure of various seeds, including the soybean, has also (117) been made. The Bureau of Plant Industry has taken a leading part in this agronomic development of the soybean (27, 72, 73) which has extended from the Southern (74) to the Northeastern States (114), as well as through the Middle West. Some of the State agricultural experiment stations have also been active in this work, among them Alabama (4), Arkansas (22), Delaware (41), Illinois (23, 24, 25, 43, 92, 96), Indiana (84), Iowa (70, 112), Kansas (105, 119, 120), Maryland (69), Massachusetts (1, 15), Michigan (66), Minnesota (11), Mississippi (12, 80, 81, 118), Missouri (34, 35, 36, 56, 88), Nebraska (54, 55), New York (113), North Carolina (116), Ohio (9, 18, 106, 107), Pennsylvania (77), South Carolina (17, 44), and Virginia (99).

Very interesting and important results have been obtained on the reaction of the soybean plant to different fertilizer salts. For instance, the amount of protein in the bean may be governed, within limits, by the fertilizer used (3, 6, 39, 75), or the growing conditions (53, 103). Effect of intensity of light and length of growing day (19, 82, 83), influence of space and arrangement of plants on production (115), effect of age on vitality of seed (59), and carotene content (97), have also been studied. A number of articles have been published on the economic status (42, 104) of the soybean, a very necessary phase to be considered and one too frequently overlooked in a new development. The possibility of industrial applications of soybean meal has been mentioned in the literature in a general way (29, 33, 48, 49, 50, 52, 78, 79) by a number of writers. Some of these articles are merely popular reviews of earlier work (48, 49, 50). The Farm Chemurgic Council (13, 14) and one large manufacturer (21) have been very active in the development of industrial applications for soybean meal, and their accomplishments are published in the proceedings of the Dearborn Conferences (78).

In 1936, under the Bankhead-Jones Act, the U. S. Regional Soybean Industrial Products Laboratory (57, 58, 68, 71) was established and organized at Urbana, Illinois, for the purpose of ascertaining the effect on the chemical composition of the soybean, of varietal, climatic, soil, and fertilizer differences, and developing new industrial uses and improving present industrial outlets for soybeans and soybean products. One section of this laboratory has devoted its entire time and effort to the study of the chemical characteristics of soybean meal with a view to developing industrial applications for the protein. The results obtained from this work are outlined in the proper sections of this paper.

TREATMENT AND PROCESSING OF WHOLE SOYBEANS

The treatment of the bean, storage conditions, and methods used for the extraction of the oil have very important effects on the protein and determine its value for most industrial applications. Various factors, such as the effect of atmospheric conditions on the moisture content (142), storage conditions (147, 166, 199), methods of preservation (122, 125, 165), bleaching (155), general treatment (133, 163, 164), suitable drying equipment (123, 124), and means of preventing or minimizing dust explosions in soybean processing plants (156) have been studied by a number of investigators. One of the several machines for producing proper flakes for solvent extraction is described (149).

Up to the present time soybean oil has been relatively more important commercially than the meal. Several methods have been used (137) for the removal of the oil, among which may be mentioned hot and cold presses of various types, hydraulic (140), expeller presses, and solvent extraction.

Manchurian soybeans are processed largely in either hand-screw or hydraulic presses. Early in the development most of the domestic soybeans were processed in expeller presses, but the solvent extraction process is increasing in application and, as industrial uses for the meal or protein expand, this trend will probably be accelerated. As shown later in this paper, the solvent-extracted or oil-free meal is a better material for most industrial applications than are the expeller meals or press cakes.

Some processes have been suggested that are concerned with the separation not only of oil and meal but also of lecithin (145, 146, 150, 151, 152). A number of processes and different lay-outs of equipment (126, 132, 136, 138, 139, 141, 144, 148, 153, 154, 157, 158, 160, 161, 162) are used in extracting the soybean oil with solvents. The usual solvent used is a light petroleum naphtha, such as hexane, but ethyl alcohol (143, 159), alcohol and benzol (127), and chlorinated solvents (135, 160) have also been proposed. The removal of the last traces of solvent from the meal (128) and of odor by pretreatment with sulfur dioxide in aqueous solution (129, 130, 131) or ammonia (134), has also received attention.

PROTEIN EXTRACTION FROM SOYBEAN MEAL

The extraction of protein from soybean meal with water to make soybean milk has been practiced in the Orient for many years. Some industrial processes utilize this method of extraction (176, 184, 215, 216, 217, 253, 254, 255, 256) which is sometimes followed by coagulation of the protein with a suitable agent (179, 219, 242).

Meissl and Böcker (212) were apparently the first to investigate the properties of the nitrogenous constituents of the soybean. They extracted the protein with water, sodium chloride, and potassium hydroxide and coagulated it with acids. The next important investigation was conducted by Osborne and Campbell (226) who extracted the protein by means of salt solutions and precipitated it by dilution, dialysis, or the addition of ammonium sulfate. Among the several fractions obtained, the principal one was a globulin which they called "glycinin." Several other investigators (174, 183, 187, 204, 248, 257) separated glycinin by using a procedure similar to that of Osborne and Campbell and studied its properties. Ryndin, et al. (229), found two fractions of glycinin possessing two different viscosities and osmotic pressures in dispersion. Hartman and Cheng (186), Jones and Gsonka (198), and Kondo, et al. (202), modified Osborne's procedure. Patents have been obtained to cover the extraction of protein from soybean meal with aqueous salt solutions or sea water and its recovery by acid coagulation (168, 169, 170) or dialysis (171, 172, 173, 185).

An early extensive study of soybean protein extraction (233, 234, 235, 241) and industrial utilization (508, 509) was made by Satow using

dilute aqueous caustic or alkaline salt solutions (236, 240) and water (237, 238, 239), but sodium sulfite (234, 235, 240) was finally recommended as the best reagent. He coagulated the protein from its extracts with such acids as sulfurous, phosphoric, sulfuric, acetic, hydrochloric, nitric, and lactic. He also investigated the use of the following coagulating agents: Alkaline earth salts, heavy metal salts, formaldehyde, ferments, dialysis, and heat.

Other workers, conducting studies similar to those of Satow, used alkalis and alkaline salts extensively (177, 178, 181, 182, 183, 191, 192, 194, 196, 197, 227, 228, 244, 258). Neutral salts were used in several investigations (191, 230, 243). Hartman and Cheng (180), Satow (237, 238, 239), Nagel, et al. (220), Woodruff, et al. (261), Ryndin (230), and Smith, et al. (243), used water as the protein extracting agent. Horvath (189, 190) discussed the chemistry of soybean protein extraction. Smith, et al. (243), at the U. S. Regional Soybean Industrial Products Laboratory, found by investigating the peptizing action of various salt solutions on protein in oil-free soybean meal ground to pass through a 100-mesh screen that distilled water was a better dispersing agent than neutral salts in concentrations up to 2 N, and that at very low concentrations salts greatly inhibited the dispersion of the protein. This is clearly shown in figure 1, page 6, as is also the fact that salts with divalent cations in very dilute solutions exert a much stronger inhibiting action than do those with univalent cations.

Smith and Circle (244) extended the investigation of peptization of soybean protein from oil-free meal to include a study of the effect of pH of the extracting solution, both with and without salts. Some of these results are shown in figure 2, page 7. It should be noted that the point of minimum dispersion is pH 4.1, thus indicating that this is the isoelectric point of soybean meal. The same investigators (245) studied the recovery of soybean protein from various dispersions by means of acid coagulation and electrodialysis. The point of maximum protein recovery is pH 4.1, as noted from figure 3, page 8.

It has long been known that the properties of vegetable proteins, such as dispersibility and viscosity, are affected by heat, mechanical treatment, and contact with various organic solvents used in the removal of the oil. This phenomenon is loosely termed denaturation. The denaturation of the protein in the whole bean and in the meal was studied (249) as a function of its solubility after heating, freezing, boiling, and treating with gasoline. Nagel, et al. (221), studied the dispersion of protein from soybean meal with alcohol-water mixtures of all proportions. Likewise, Okano and Ninomiya (224) and Mashino (191, 208, 209, 210, 211) investigated the effect of alcohol treatment on the dispersibility of the protein. Chlorinated solvents were also studied (135).

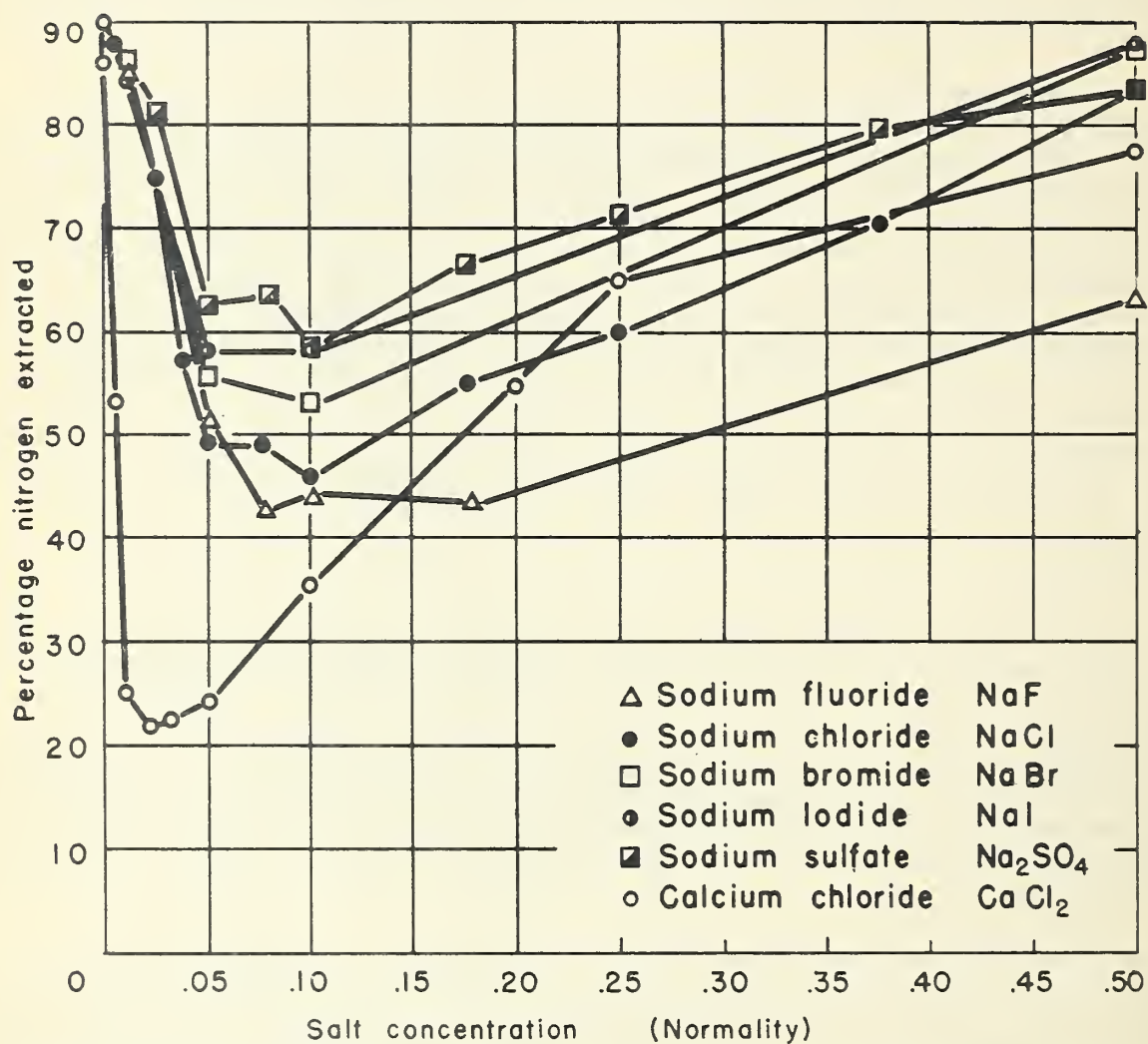


Figure 1.— The total nitrogen dispersed from solvent-extracted soybean meal by various salts

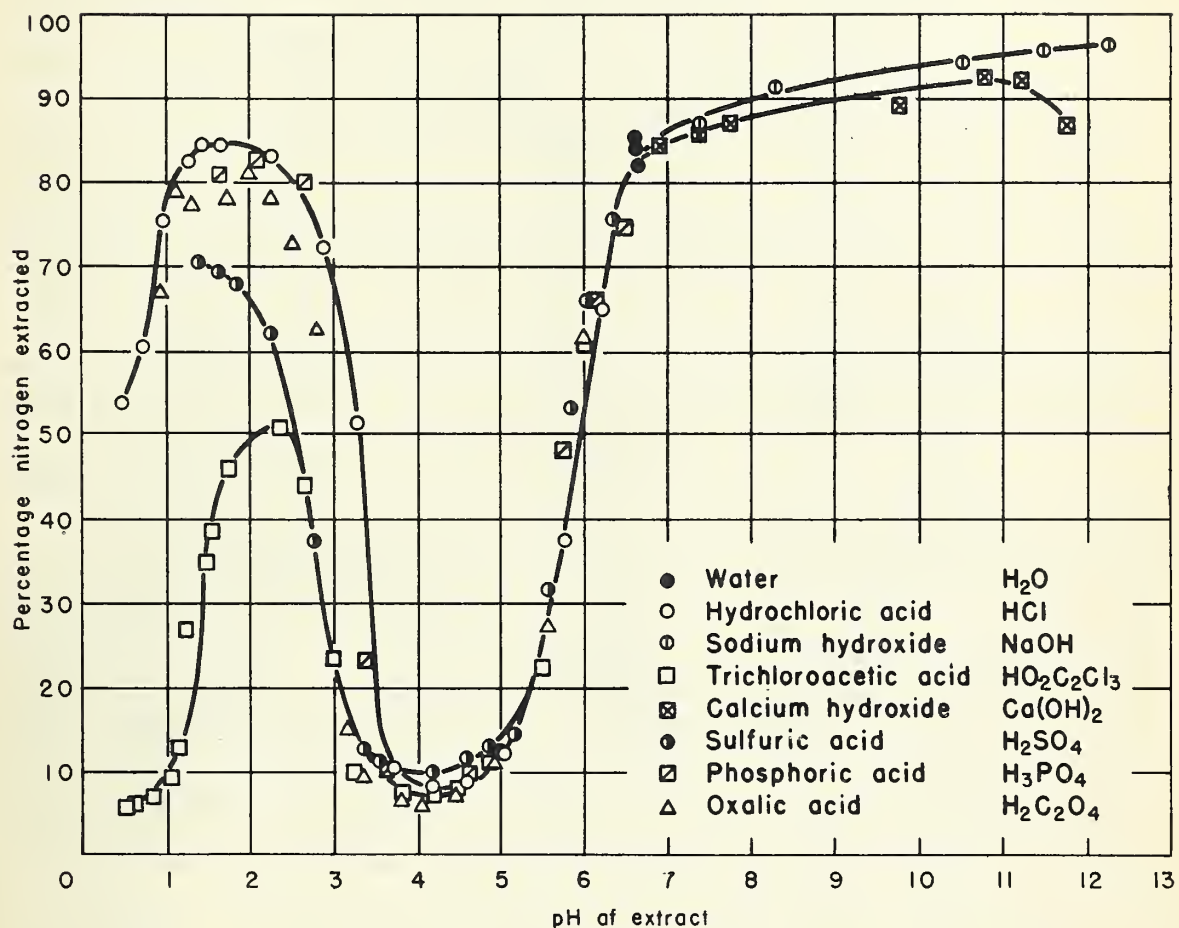


Figure 2.- Percentage of total nitrogen extracted from oil-free soybean meal by various acids and bases

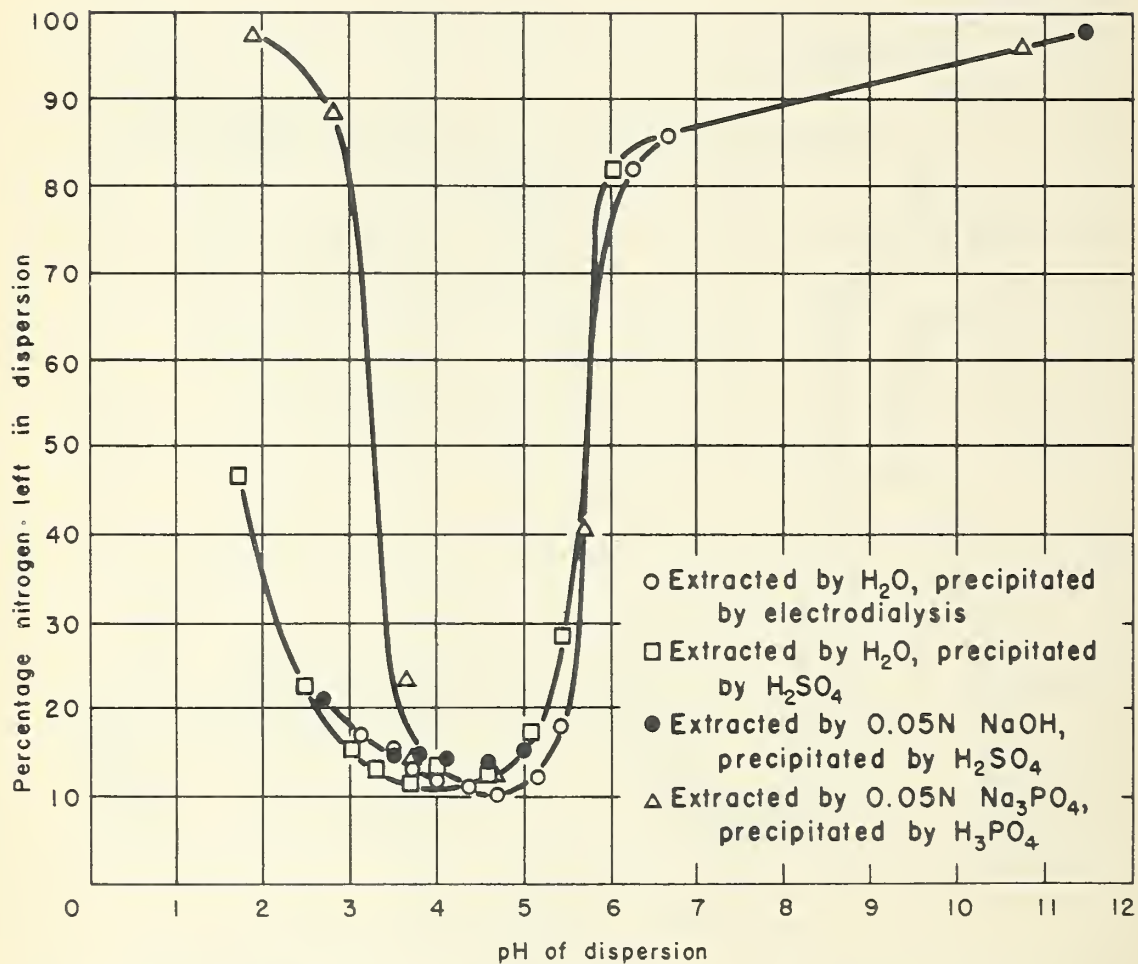


Figure 3.- The precipitation of soybean protein from water and alkali dispersions by various acids

The effect of aging soybean meal on the dispersibility of its protein in salt solutions was studied by Jones and Gersdorff (199). A similar study, with emphasis on the dispersion of protein in water, was made by Smith and Circle (244). Nagel, et al. (220), investigated other physical factors affecting the dispersion of soybean meal in water.

The bound water relationship (203) and methylation (200, 204) of soybean protein were studied. The isoelectric point of glycinin was determined (187, 214) to be at pH 5.0, whereas Monaghan-Watts (213), investigating the whipping characteristics of soybean meal dispersions, found the isoelectric point of oil-free soybean meal to be pH 4.1. The heat of combustion of glycinin, which is claimed by some investigators to be combined with 5.1 percent carbohydrate (257), was determined as 5,668 calories per gram (174). The speed of racemization (205), formation of "proto acids" (183, 227), and various other properties (195, 206, 207, 218) of soybean protein were investigated and certain properties compared with those of casein (252). Factory methods of equipment used in Russia (250) for producing soybean protein were described and illustrated. Koide (201) recovered soybean protein from its dispersions by spraying it into hot air chambers.

A procedure used for modifying the properties of soybean protein and for rendering it more readily dispersible is by treatment with enzymes (251). Pepsin (175, 231, 232), papain (222), and trypsin (167, 223, 232) have been used in this connection and their actions, as well as those of certain fungi (225, 259, 260), have been studied.

One of the most important reactions in the industrial use of protein materials is that known as hardening, tanning, or curing. The conversion of raw collagen into leather and of casein into casein plastic material are well-known examples. It appears that the inorganic salts, such as those of chromium, aluminum, and iron, or organic materials, such as tannic acid and aldehydes, especially formaldehyde, are the best agents. It has been claimed (193) that pretreatment with methyl alcohol renders soybean protein more reactive toward formaldehyde. Actually, there is very little known about the chemistry of this important reaction. The U. S. Regional Soybean Industrial Products Laboratory started to investigate fundamentally the soybean protein-formaldehyde reaction, and Smith, Max, and Handler (246) found it possible to disperse protein from soybean meal in an aqueous solution of formaldehyde. Further work (247) has been done to make possible the preparation on a commercial basis of this dispersion, since there will doubtless be many industrial applications made with it. Some of the more obvious applications are mentioned later.

HYDROLYTIC PRODUCTS OF SOYBEAN PROTEIN

Soybean protein is given hydrolytic treatment for two purposes, namely, to modify the protein for certain industrial applications and to

determine the break-down products, such as the amino acids. The hydrolytic treatment of soybean protein for the purpose of improving its properties as an adhesive or plastic has been studied in an empirical way (182, 247, 479, 509, 512).

The hydrolysis of soybean protein has been carried out with acid (285, 286), alkali (285), superheated steam (283, 308), and enzymes. Enzymatic hydrolysis has been discussed in the preceding section (167, 175, 222, 223, 231, 232, 251, 259, 260). (Reference is also made to 282, 304, 306.) The conditions and rate of hydrolysis have been studied in a general way (285, 286, 309), and electrolytic oxidation of crude protein hydrolysate has been carried out (311).

The nitrogen distribution in soybean protein and meal has been studied by several workers (271, 273, 289, 296, 305), using principally the methods of Van Slyke. The effect of successive reprecipitations of glycinin on the distribution of nitrogen in its structure has received some attention (284).

Mild hydrolysis leads to intermediate products such as metaproteins, proteoses, peptones, and polypeptides (274, 283, 308). More complete hydrolysis yields the amino acids. The hydrolysis of glycinin has yielded the following amino acids (262, 272, 279, 287, 288, 295, 301): Glycine, valine, alanine, leucine, isoleucine, proline, phenylalanine, tyrosine, cystine, aspartic acid, glutamic acid, arginine, histidine, lysine, and tryptophane. The following amino acids have received more attention than others: Proline (307); the basic amino acids, lysine, arginine, and histidine (270, 280, 281, 293, 298, 299, 316); cystine (266, 267, 275, 313); methionine (313, 314); tyrosine (266, 267, 303); tryptophane (266, 267, 291, 313); aspartic acid (293); and glutamic acid (265, 290, 293, 297, 310). It is claimed (293, 294, 315) that hydroxy-glutamic acid has also been isolated from glycinin.

A comparison of several varieties of soybeans with respect to amino acid content has been made (266, 267, 277, 278). The amino acids of the soybean have also been compared with those derived from other vegetable seeds (271, 273, 276, 279). A study has been made of the distribution of amino acids in the protein of the soybean embryo (292), and biological changes occurring in the proteins of soybeans during germination have also been investigated (264, 269, 300, 302, 312).

Other nitrogenous constituents of soybean meal obtained on hydrolysis are the organic bases, adenine, guanine, choline, asparagine, trigonelline, putrescine, and cadaverine (268, 298, 299, 316, 317). Some of these bases are decomposition products of the amino acids caused by drastic conditions of hydrolysis, while others are probably derived from nucleoproteins (263) and phosphatides.

ENZYMES OF THE SOYBEAN

Several publications deal with the identification of the various enzymes which occur in the soybean. Those enzymes which have been reported are oxidase, protease, and urease (374); amylase, glucosidase, protease, peroxidase, lipase, and urease (387); catalase and nuclease (349); purinoxidase, uricase, allantoinase, allantoicase, and urease (330); allantoinase and uricase (335); glyoxalase (358, 382); phosphatase (336); catalase, amylase, and protease (361); and antioxygenase (340). A general summary of the subject has been made by Waksman and Davidson (397).

The urease enzyme in the soybean has received the most study. The first reference to its occurrence seems to have been made by Takeuchi (388). The distribution of urease in the soybean plant and other legumes has been investigated (384). It has been compared with the urease from Robinia (367) and that from the Canavali or jackbean (338, 342, 399, 402). Also, comparison of urease content of several varieties of soybeans has been made (329, 342, 400, 402). It was found in the roots, stems, seedpods, and leaves of young plants as well as in the seed (403), the outer epidermis of the cotyledon (396), the cotyledon (338), and the parenchyma cells of seedlings (339).

Urease was usually prepared from a water or glycerol suspension of soybeans (380). According to some investigators, it was not very stable in water dispersion, but in the form of a powdered concentrate it kept for a considerable period of time (345, 348, 393, 406). However, Wester claimed that a water dispersion retained its activity for several months (399). Urease was concentrated by precipitation from solution with an antiurease developed immunologically (357, 387).

The existence of urease in soybeans was questioned and the conversion of urea to ammonium salts ascribed to bacterial action (365), but this point of view was later refuted (328, 341, 398). The activity of urease in soybeans was not diminished by their storage for several years (329, 398, 400, 402), and claims were made that aging of the bean increased its urease activity (395).

The action of urease in soybeans was believed to depend on the presence of a coenzyme which was dialyzable and claimed to be thermolabile (375). Kato, however, believed that the coenzyme was thermostable and that the enzyme itself was destroyed by heat (352). The urease activity increased on warming to 37° C. and then decreased with time. Wester believed that this suggested the hydration of the enzyme or a more active intermediate enzyme (401). Glycine was supposed to act like the coenzyme in augmenting the urease activity (351, 354, 355). It was claimed that phosphates inhibited the activity of the coenzyme (353), while another author believed that they were without effect (368).

Takeuchi (388) stated that 1-percent sulfuric acid, 1-percent hydrochloric acid, 1-percent sodium hydroxide, 5-percent ammonium sulfate, 0.05-percent copper sulfate, 0.25-percent sodium fluoride, and 0.05-percent mercuric chloride inhibited the activity of urease from the soybean to some extent, but doubling these concentrations resulted in complete inhibition, while magnesium oxide even in large quantities had no adverse effect. The inhibiting action of acids and bases was further investigated (363). Other inactivators or poisons investigated were nickel oxide (347), thioglycols and mustard gas (381), and ferrous sulfate, sodium thiosulfate, and hydrogen sulfide (359). The lime liquors used in dehairing hides inactivated urease and other enzymes (327). Bicarbonated mineral waters were said to contain both activators and inactivators for soybean urease (362). Gum arabic and starch appeared to augment its activity (390). Monochromatic light also accelerated urease activity (366), whereas direct sunlight was supposed to be deleterious (398).

Labberte (359) studied the action of a water extract of soybeans on urea by titrating the quantity of ammonia formed. This enabled him to determine the various factors influencing the reaction. Wester (399, 404) also studied the quantitative relationships between urea and urease. Van Slyke and Cullen stated that the concentration of the urea solution could be varied between 0.2 and 10 percent without influencing the rate at which ammonia was formed by a given concentration of urease. They also found that excess of ammonia retarded the rate of reaction and that between 10° and 50° C., each 10° rise doubled the rate, the optimum temperature being about 60° C. (392, 394). The urease of the soybean was used in a qualitative test for urea (377). It is interesting that the action of urease may be reversed. Kay (356) claimed to have formed urea from highly concentrated solutions of ammonium carbonate and carbamate by its use.

The injection of soybean-urease solutions into the blood stream of a dog proved fatal, accompanied by an increase of ammonia in the blood (324, 325). Methods were described for differentiating the action of urease from that of uricase and albumins in the blood stream (379). Van Slyke's procedure of estimating urea with soybean urease was used by Eigenberger in a study of several normal and pathological urines (331). The activation of soybean urease by human serum (371) and its distribution in normal and previously treated organs were investigated (333).

Several workers have studied the proteolytic enzymes found in soybeans. They were primarily interested in comparing the relative proteolytic activity of extracts of soybean seeds, soybean sprouts, and seedlings (378, 385, 389). The soybean proteases were also compared with those from seeds of other plants (323, 378).

Several papers have been published which deal with the starch-splitting enzymes in soybeans (321, 337, 350, 364, 376, 383, 391). Newton and Naylor

(372) described methods for determining the saccharogenic power of soybeans and soybean-amylase concentrates. Ascorbic acid oxidase (346) and a β -galactosidase (343) were found in the soybean.

The catalase and urease content of soybeans was studied with respect to germinative capacity (370), and it was reported that for seed treated under comparable conditions the activity of the catalase as measured by the quantity of oxygen evolved may be a convenient method for determining in a few minutes their germinative capacity. The chemical kinetics of soybean catalase was investigated (405), and it was reported that catalase was a common name for a group of enzymes having different stabilities and different physical properties. An oxidase (373), a peroxidase (326), and an antioxygenase (340) were also investigated as constituents of the soybean.

In 1920 uricase was reported to be present in soybeans (369), but another investigator (360) in 1927 was unable to find evidence of its presence. However, in 1930 two procedures were described (334) for destroying the uricase activity in soybeans without affecting the activity of the allantoinase and urease.

Falk (332) compared soybean lipase with that of castor bean lipase and concluded that they were very similar. Barton (322) also compared soybean and castor bean lipases and inferred that they were the same except that soybeans contained more than one lipase. The presence of fat oxidases in the soybean has been reported (318, 319, 344), and their properties have been compared with those of enzymes found in the kidney bean and hyacinth bean (320).

INDUSTRIAL APPLICATIONS

Adhesives

Adhesives with protein bases have been known and used successfully in industry for many years. Bone, hide, fish, and casein glues are well established in the wood working industries and a new product would have to be materially better or cheaper to find an opening. It is claimed (455) that in 1932 soybean meal (9 years after its introduction) was used in the American plywood industry to an extent equal to all other adhesives. This industry consumes an average of 17,000 tons annually, but it has been claimed that as much as 25,000 tons have been consumed. The bulk of this meal has been used in the far West and consists largely of imported Manchurian oil cake. Hadert (442, 443) published review articles from which a more general picture may be obtained.

Soybean meal adhesives are so recent in their development that a number of questions have been raised regarding price, spread, and

workability (407). Comparisons have been drawn between soybean protein and casein (446, 468, 473), as well as between the meal and the commercial soybean "alpha" protein (411). Adhesives have also been prepared from soybean refuse (449).

In general, it would appear that the solvent-extracted soybean meal or the commercial "alpha" protein would be the best material for the preparation of protein adhesives. Meal ground to pass through a 200-mesh screen (423) has been found suitable. Also, in the preparation of adhesives, the meal may be improved by subjecting it to a preliminary treatment with such agents as weak acid (440) to remove carbohydrates, an enzyme (479), alkaline earth peroxides (438), or other agents (439), and extracting the oil at low temperatures (469). In preparing adhesives an aqueous dispersion of the soybean meal or protein is usually made with dilute caustic (452), but sodium or ammonium carbonate (444), borax (422), or an aqueous solution of urea (408) may be used. In order to increase the water resistance of the adhesive, lime is frequently added to the caustic dispersion, and other salts, such as sodium sulfite (418), an alkaline silicate (409, 445, 453, 456, 459, 464) or fluoride (425, 426, 450, 475), zinc chloride (458, 462, 478), sodium dichromate (466), a copper salt (456, 459, 460, 465), a manganese salt (454), or an organic salt of aluminum, ammonium, barium, or calcium (414), may be added. Sulfides, principally carbon disulfide (425, 445, 453, 457, 459, 465, 467), rosin (409, 420, 426, 454, 456, 460, 475), carbon tetrachloride (420, 453), chloroform (420), Portland cement (463, 465), latex (447, 448), phenol, or naphthol (426, 434, 462) may be incorporated for the same purpose. Phenol, naphthol, or cresol may be mixed with the protein dispersion, to be followed by the addition of formaldehyde (441, 471, 476, 477, 480), phenol-formaldehyde resin (470), or urea-formaldehyde (412).

The soybean glue may be mixed with starch glue (415), starch viscose (413), or cellulose viscose (451, 474). It may be mixed with a casein dispersion (410, 416, 417, 419, 467, 482), or casein with 30 to 35 percent of water, and extruded from a die (437). It may also be mixed with albuminous material (435) or powdered scrap vegetable ivory (436). It has been proposed as a binder for briquetting coal dust (475).

In order to improve the water resistance of the adhesive, it has been proposed to add protein tanning agents, such as quebracho (456, 462), tannins (465), or chromium or aluminum salts (461). However, it has been recognized that aldehydes are the best waterproofing agents, and attempts have been made to prepare adhesive mixtures with various aldehydes (472), furfural (409), and formaldehyde, either in very small quantities (447) or in the presence of retarding agents, such as ammonia (481) or a mixture of blood and caustic (421). A formaldehyde jelly (424) has been proposed for panel veneer gluing.

To prevent the wood from staining, the quantity of water in plywood adhesives is reduced by using dry soybean flour sprinkled on wet wood surfaces (427, 428, 429, 430, 431, 432, 433) which are united by applying pressure with or without heat. The soybean meal is sometimes used alone and in mixture with a water-soluble alkali in dry form.

The soybean protein dispersion in formaldehyde (247) already mentioned should find quite extensive application as a water-resistant adhesive.

Plastics

Casein from milk is the principal protein material used today in the production of plastics. Because of certain apparent similarities in behavior and because its cost as a byproduct (502) would be low, soybean protein has recently been proposed (483, 514) as a substitute to at least partially replace casein. Furthermore, soybean protein is finding application as a plastic in fields practically untouched by casein (516). The development of soybean plastics has followed somewhat different lines in the United States (501) as compared with other countries.

The early development of plastic material from soybean protein closely followed that of casein. At first, mixtures of the soybean meal or protein acted as binding material (505) without any attempt being made to harden it. Then the wet-process methods were introduced, first without hardening (493, 504), then with formaldehyde hardening (495, 496, 498, 506). The wet process for protein plastic production consisted in dispersing the protein, usually in an aqueous caustic solution, coagulating the protein with acid, a salt, or a hardening agent such as formaldehyde, and pressing the wet curd. Satow, the investigator who contributed so much to the early study of the extraction of protein from soybean meal, took the lead in the early investigation of the possibility of soybean protein for plastics. At the time he conducted his investigations on the soybean, the dry process of manufacture of casein plastic material was not generally known, so it is to be expected that Satow's work (508, 509, 510, 511, 512) was on the wet process. He further specified that the protein must be glutinized by treatment with strong acid or alkali before the formaldehyde treatment. This glutinization produces a material which has excellent plastic flow and gives a beautiful molded piece when it is first removed from the die, but on standing a few weeks the piece will spontaneously fracture and shatter. Horvath (49) recently outlined a process for the production of soybean protein plastic material by a dry process, but his method is merely a review of Satow's work with an attempted adaptation of Satow's work to the production of a dry molding powder. No experimental data were given, other than those previously published by Satow.

Satow (513) employed soybean meal, together with blood albumin, as a binder for a fibrous material in the production of wallboard. Soybean meal has also been suggested (519) as a binder for clay in the preparation of crayons. A rather unusual wet process for plastics is the use of a soybean dispersion mixed with an aqueous dispersion of cresylic acid (507) and formaldehyde and also with soap and aluminum acetate (515) followed by an aldehyde.

The only commercially successful application of soybean meal to the production of plastic material that has been made to date is the well-known Ford development (492, 494, 497, 500, 503, 506, 517). Here the meal is mixed with wood flour, phenol, formaldehyde, lime, and other materials and processed to form a molding powder. The proportions are roughly: 25 percent soybean meal; 25 percent wood flour; and 50 percent phenolic resin. Even when used in this comparatively small amount and in the untreated state, there is little doubt that the soybean meal reacts to some extent with the formaldehyde and possibly with the phenol, so that it fulfills more than the function of merely filler material. Mixtures of soybean protein with resins have also been tried (518).

When work on the investigation of possible plastic applications for soybean meal and protein was initiated at the U. S. Regional Soybean Industrial Products Laboratory, it was proposed to approach this problem from an angle somewhat different from that usually employed in work on protein plastics. Instead of studying the protein plasticized with water, a study was made (484, 499) of plastic flow at reduced water content. This study was preliminary to attempting the production of a molding powder that would allow molding a protein plastic that would come finished from the die. It was obvious that this would not be possible if plasticization required the presence of excess moisture. Next it was found (487) that soybean protein, hardened or tanned with formaldehyde at a pH of 4.1, produced a material that was thermoplastic and of minimum water absorption. Figure 4, page 17, shows how the water absorption increases in soybean protein hardened in formaldehyde at either below or above pH 4.1. The formaldehyde-hardened soybean protein material, although thermoplastic, requires 10 percent or more water content and rather high molding pressures. However, it was found that this powder could be prepared with 5 percent or less moisture content and plasticized with ethylene glycol (488), glycerol, ethylene cyanhydrin, furfuryl alcohol, or the like. None of these can be considered entirely satisfactory, but they do permit the protein plastic to be molded to shape into a piece which comes finished from the die. It is claimed that ethyl protein-formaldehyde (485) is also thermoplastic.

However, since formaldehyde-hardened soybean protein material is thermoplastic, it must be chilled in the die under pressure before removal and yet it does not flow readily enough to allow molding in injection dies.

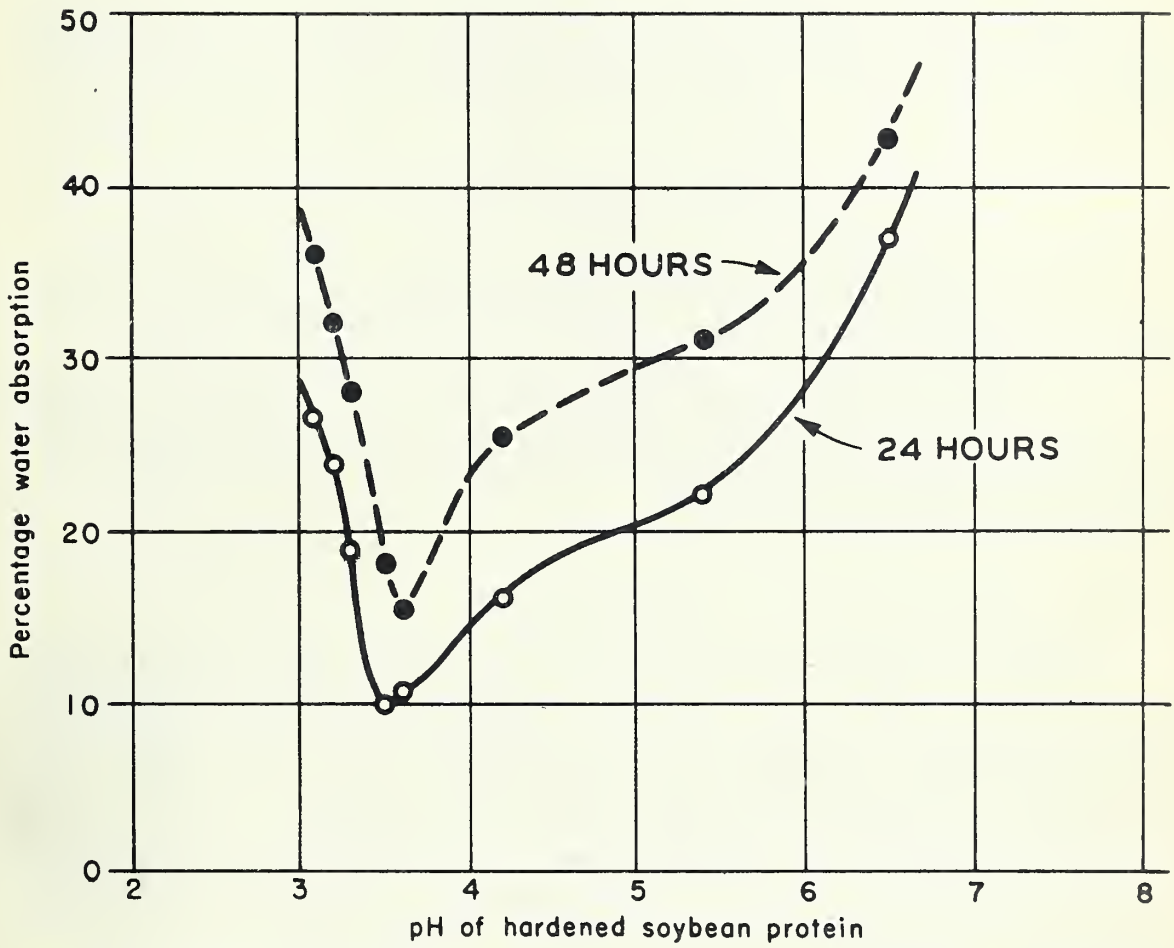


Figure 4.- Variation of water absorption of formaldehyde hardened soybean protein with change in pH

To be of commercial importance, it must be made to work in injectors or be rendered sufficiently thermosetting to allow removal from hot dies. It was found that the formaldehyde-hardened soybean meal or protein was compatible with phenolic and urea resins (490), so further work was done with the soybean meal-phenolic resin (489) mixtures and some rather promising results were obtained. The soybean-urea resin mixtures also invite investigation. Considerable work of a fundamental nature (491, 497) remains to be done, results of which cannot be predicted (486) at this time.

Another line of development of considerable promise is the application of the soybean protein-formaldehyde dispersion (247) to the production of plastics. The soybean protein in this dispersion is uniformly hardened protein salt which on drying becomes thermoplastic (487). Unsized kraft paper impregnated with this dispersion, dried to normal moisture content, and stacked and pressed, produces a laminated material with definite possibilities. Since the formaldehyde-hardened soybean protein is compatible with phenolic resins (490), it is possible to place a sheet impregnated with phenolic resin on either exposed face, with the result that the laminated material will have the water resistance of phenolic material. There should be a large field for such a material in the building industry.

The soybean protein-formaldehyde dispersion may be used to impregnate any fibrous material such as tanned leather scrap. This impregnated material, when dried, gives a very promising product from which very cheap but good plastic materials may be made.

Paper Sizing

Casein is used very extensively for the coating and sizing of paper. According to a recent estimate, 75 percent of the casein used in American industry is used in the paper industry. Soybean protein has some characteristics which make it suitable for this application, but others, such as its darker color, have prevented its use to any large extent. Some work on the investigation of pigments in soybean meal has been reported (537, 542), but no practical method has yet been proposed to improve the color sufficiently to make it suitable for high-grade paper coatings. Since the coating requirements for wallpaper are less exacting than for writing and high-grade printing paper, soybean meal and protein have been adapted to this and similar applications (520, 522, 523, 535, 543).

Several patents have been granted for the use of soybean meal or protein as a beater size (521, 525, 527, 529, 530, 531, 532, 533, 534, 538, 539). Rowland (539) prepared a size by dissolving soybean protein in saponified rosin and adding boric acid. This gives a very stable dispersion of free rosin with an unusually small particle size. Kress and Johnson

(530, 531, 532, 533, 534) stabilized a wax emulsion with soap and soybean protein. Special paper products have been prepared with soybean protein as an impregnating material (526, 540) and for slack sizing (524) prior to lacquer finishing. A subsequent hardening with formaldehyde has also been developed. Soybean protein in sizes for textiles has also been proposed (528, 541).

The soybean protein-formaldehyde dispersion (247) should find wide application in the finishing of paper. It is understood that over a ton of soybean protein per day in the form of this dispersion is now being used for sizing glassine paper, and other developments along this line are known (536). This application will doubtless increase as the dispersion becomes better known.

Miscellaneous

As was the case some 20 years ago in the plastics field, the development of synthetic fiber from soybean protein is today paralleling that of lanital or casein-wool (555, 560) from casein. It has been reported (544, 545, 554) that the Japanese are building a factory to produce from 20 to 30 tons daily of synthetic wool fibers from soybean protein and that the Italians have experimented with soybean protein (548) and are preparing to grow soybeans in Ethiopia (546) to mix with casein in making lanital. In the United States, the Glidden Company announced the production of a fiber from soybean protein (547). The usual method is to extrude the properly treated soybean protein dispersion, containing stabilizers, such as sugar, tartaric acid (556, 557), lecithin (558, 559), or urea (549, 552), into an acid bath to be coagulated and then through a formaldehyde bath to be hardened. If fixing agents, such as borax, phenol, or creosote are used, formaldehyde treatment is claimed to be unnecessary (553). Mixtures of resins and vegetable protein, such as soybean protein, have also been proposed (550, 551). It is a much debated question just how important this development is to America where there is no shortage of natural wool, and natural wool is stronger than these synthetic fibers (561), but whether of immediate practical importance or not, the development is of considerable interest and well worth following.

In the preparation of films from soybean protein, formamides (562) or a mixture of boric acid and a polyhydric alcohol (563) have been suggested for use as restraining agents for formaldehyde. This is another field where the soybean protein-formaldehyde dispersion (247) can probably be applied to advantage.

Soybean protein has been mentioned for use in cold-water paints (564, 565, 566, 567) of varying formulation and with various claims, few of which

can be supported. The soybean protein-formaldehyde dispersion (247) will probably give as satisfactory paint of this type as is possible to prepare, but it will not be comparable to drying oil paints.

A number of miscellaneous applications for soybean protein or meal have been suggested, among which are its uses as a dispersing or emulsifying agent (568, 569) for mineral oils or fats, stabilizing agent to inhibit rancidity in fats (570, 571, 572, 573, 574) or glue (575), bleaching agent for wheat flour (576, 577, 578, 579, 580, 581, 582, 583), adhesive in insecticidal sprays (584, 585), core binder (586), clarifying agent for tanning extracts and dyestuff solutions (587, 588), mellowing agent for aging alcoholic distillates from grain (589, 590), component of a polishing or finishing wax (591), and wetting-out agent (592). There are doubtless numerous other possible applications which will be attempted in the near future, many of which will be successful.

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TREATMENT AND PROCESSING OF WHOLE SOYBEANS

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INDUSTRIAL APPLICATIONS

Adhesives

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